COHESIVE LAWS IN ADHESIVES JOINTS: THE TEARING/DEBONDING TEST FOR CHARACTERIZATION OF THIN ADHESIVE FILMS

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ABSTRACT

To be able to predict the strength of adhesive joints accurately, correct material data of adhesives are essential. Hence, it is critical to develop reliable testing methods to obtain the constitutive behaviour of adhesive layers. In use, adhesives are constrained to thin layers. Thus, an adhesive constrained into a layer is expected to behave differently compared to the adhesive as a bulk material. Under loading, the size of the Failure Process Zone (FPZ) in the adhesive layer is often much larger than the thickness of the layer. Thus, the small scale FPZ condition is not fulfilled and the traditional Linear Elastic Fracture Mechanics (LEFM) can not be applied. At the same time, experiments show that test specimens are prone to produce unstable crack propagation and combined adhesive/cohesive fracture patterns appear frequently, especially when mixed mode loading (peel and shear) is involved. Cohesive law should be taken as the basic fracture property for adhesives characterization; cohesive laws must be determined experimentally. The effects of loading rate and adhesive layer thickness on the cohesive law shape have to be investigated experimentally. The coupling of elasticity, adhesion and fracture make difficult interpretation of test results, especially if the adhesive is an elastomer, which has a failure strain of several hundred percents. A new test has been proposed, combining tearing of the adhesive layer and debonding from the substrate in a controlled way and using a simplified geometry. Results are closely related to the stiffness, work of fracture and adhesive energy of the adhesive system, all of them plying simultaneously an active role during the very same test.

KEY WORDS: adhesive, tearing, debonding, cohesive law.

1. INTRODUCTION

Some of these mechanisms of energy absorption and toughness improvement both in biological and bioinspired materials are:

1. Rupture of “sacrificial” weaker bonds in the macromolecular component.
2. Extension, pull-out and/or ligament formation of a macromolecular component bridging an interface
3. Void formation leading to bulk plastic deformation, crack blunting, pinning and branching.
4. Localized plastic deformation ahead of a crack tip.
5. Microcrack formation.
6. Phase-transformations which take place ahead of a crack tip.
7. Viscoelastic dissipation.
8. Interacting nanoasperities and mechanical interlocking leading to inelastic strain.

A recent research has paved the way to develop new strategies for energy dissipation inside hybrid materials composed mainly of a granular media [1]. The goal is to employ several of these strategies altogether to obtain light and tough materials, with a high-energy absorption capability related to their low density. It is also quiet important to retain a certain residual strength after the impact to assess the structural integrity for the application in mind, that is, to assure also a good damage tolerance of the hybrid material.

In use, adhesives are constrained to thin layers. An adhesive constrained into a layer is expected to behave differently compared to the adhesive as a bulk material. The use of an un-cracked butt joint specimen to measure the constitutive relation can be tempting. However, experiments show that this specimen is prone to be unstable. The softening part of the constitutive relation, which contributes substantially to the fracture energy, is never captured experimentally. A few testing specimens and techniques for evaluating fracture properties have been developed. Most of these are based on LEFM and the flexibility of the adhesive layer is neglected [2].

However, there are difficulties with standard tests The locus of failure and the issue of directional stability of cracks in adhesively bonded joints have been investigated by different authors and, over the years, several criteria have been developed [3]. According to these criteria, a crack in an adhesive bond can be steered to different locations if the local stress state at the crack tip is in mixed mode. Consequently, various failure locations can result and failure does not necessarily occur at the weakest site within the material. T-stress plays an important role in the directional stability of the crack propagation. The crack is directionally stable if the T-stress is negative, whereas is directionally unstable if the T-stress is positive. The adherend bending effect on the T-stress induces a non negligible influence of the thickness of the adherends on the directional stability of the cracks in the adhesive joints.

Even for the most simple test specimen geometries the Strain Energy Release Rate (SERR) and /or J-integral include non clearly determined contributions of different dissipation mechanisms (fracture, debonding, viscoelastic effects). From this information, it is possible to derive the constitutive relations representing the mechanical behaviour of the entire adhesive layer. Such constitutive relations can be represented by traction-separation models, also referred to as cohesive laws, describing activities in the adhesive layer before and at fracture. It is difficult to obtain parameters that are related exclusively to only one process (interfacial debonding, fracture of the adhesive or viscoelastic behaviour of the polymer). Extrapolation to real adhesive joints is consequently limited and, specifically, there are difficulties to properly model the adhesively bonded joints in virtual testing by numerical simulation.

2. TEARING-DEBONDING TEST

A new test has been proposed, combining tearing of the adhesive layer and debonding from the substrate in a controlled way and using a simplified geometry [4]. Results are closely related to the stiffness, work of fracture and adhesive energy of the adhesive system, all of them plying simultaneously an active role during the very same test.

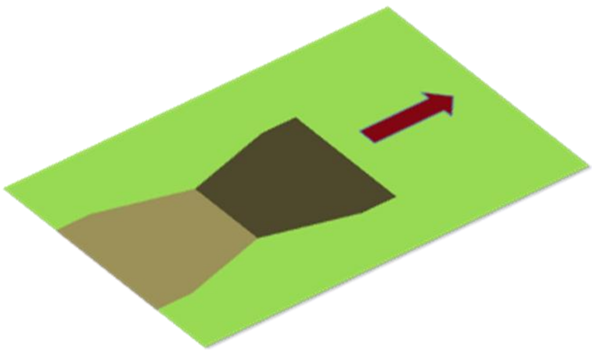

Figure 1. Simplified geometry of the tearing-debonding test.

We have all had the experience of unsuccessfully trying to remove a rectangular strip from a roll of adhesive tape by scratching the edge with a fingernail. When pulling on the partially detached piece, the strip annoyingly narrows, detaches and the final tear is often too short to be useful. Similar difficulties are experienced when trying to remove wallpaper, a sticker or a package label. The runaway tear may in fact be taking a natural physical path. Elasticity of thin sheets couples with adhesion and fracture to produce distinct shapes characterizing the tearing process. A combined experimental and theoretical study to explore this coupling of elasticity, adhesion and fracture, in a simplified geometry shows a promising way to clarify the specific role of every energy dissipation mechanism, Figure 1. We adhere a thin elastic sheet to a solid flat surface and cut two notches on one of its edges such that a rectangular flap is created, which is then pulled at a constant speed. The two crack tips (located at the edge of the flap) are initially parallel, but as the flap starts being pulled they propagate both forwards and inwards as the material progressively de-adheres from the substrate. Eventually, the two tips converge to a point and the strip detaches completely, leaving behind a triangular tear.

The profiles of three representative tears are shown in Figure 2 in which only the width of the initial flap (distance from the initial two notches) was changed. The sides of the flap are straight and make the same angle theta with the axis of symmetry of the tear, independently of the size of the initial flap.

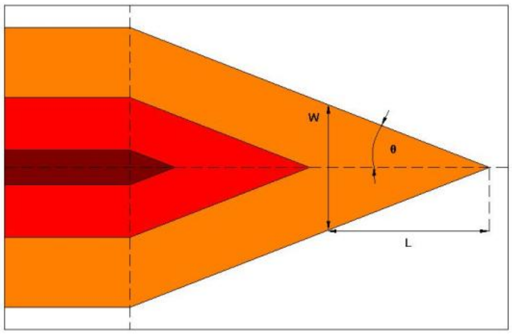

Figure 2. Profiles of three representative tears for different sizes of the initial flap.

Following Griffith's theory of fracture, a simple mechanism based on elasticity has been proposed to understand the experimentally obtained tear shapes [5]. A pulling force deforms the surface and focuses elastic energy in a fold that joins the flap with the film. This energy can be released in two ways: by decreasing its curvature (advancing the crack in the pulling direction) or by simply reducing the width of the ridge (the cracks move inwards). The actual direction is a combination of both effects, but always leads to a narrowing of the tear.

The total energy of the system that quantifies the above outlined mechanism is

$$U = U_E + 2\gamma ts + \tau A \qquad (1)$$

U_E is the elastic energy, $2\gamma ts$ is the fracture energy, and τA is the adhesive energy. The factor 2 in the fracture energy term accounts for the fact that two fracture paths are propagating along the film. The work of fracture, γ, always comes in the combination γt, and this parameter has a dimension of a force, we refer to it as 'fracture force'. Assuming that the end of the flap is always at an angle of 180° from the reference plane defined by the solid wall, we conclude that the elastic energy is only a function of the tip displacement, *x*, and the length of the strip along its axis of symmetry, *l*. The excess of length $2l - x$ is folded near the detachment line (Figure 3), so that we expect the elastic energy to be a function of the tip displacement in this combination.

$$U_E = U_E\left(2l - x, W\right) \tag{2}$$

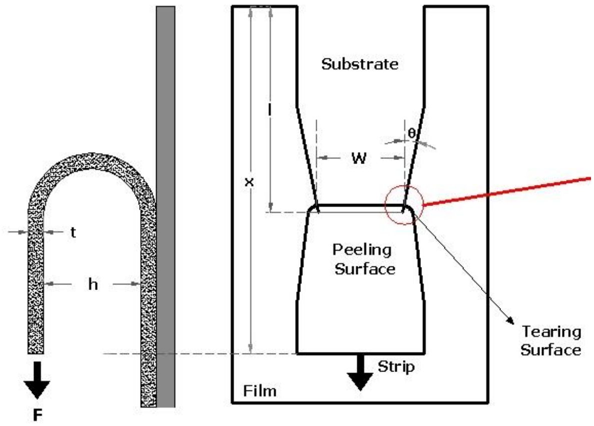

Figure 3. Folding f the adhesive film near the detachment line.

The crack tip advances to a position that minimizes the total energy. In a displacement-controlled experiment, the first variation of *U* with respect to the geometrical parameters is

$$\frac{\partial U}{\partial s} = 0$$
$$\delta U = \left(\partial_W U_E\right)_{x,l} \delta W + \left(\partial_l U_E\right)_{x,W} \delta l + \ldots$$
$$\ldots 2\gamma t \delta s + \tau W \delta l \tag{3}$$

The force is given by the work theorem as

$$F = \left(\partial_x U_E\right)_{W,l} \tag{4}$$

This equation combined with the specific dependence of the elastic energy on the geometrical parameters yields for the energy minimum

$$0 = -2\left(\partial_W U_E\right)\sin\theta - 2F\cos\theta + 2\gamma t + \tau W\cos\theta$$
$$\sin\theta = -\delta W/2\delta s \qquad \cos\theta = \delta l/\delta s \tag{5}$$

To find the fracture path, we require that the tear follows the direction where a minimal force is necessary for the advancement of the crack tips. An implicit derivative of equation (3) gives the equivalent condition usually referred to as the maximum-energy-release- rate criterion

$$\partial_\theta\left(\delta U/\delta s\right) = 0$$
$$0 = -2\left(\partial_W U_E\right)_{x,l}\cos\theta + 2F\sin\theta - \tau W\sin\theta \tag{6}$$

Equations (5) and (6) have a clear interpretation in terms of static equilibrium of in-plane forces. These forces, acting on one half of the strip, are: the fracture force (γt) resisting crack propagation, the operator pulling force *F* opposed to the adhesion energy dissipation $\tau W/2$, and the lateral elastic energy gradient $\delta_W U_E$.

The forces projected along the forward and sidewise directions give the equivalent equations

$$F = \tau\frac{W}{2} + \gamma t\cos\theta \tag{7}$$

$$\left(\partial_W U_E\right)_{x,l} = \gamma t\sin\theta \tag{8}$$

The pulling force is balanced by two forces: that of adhesion of the film to the substrate and that of fracture. It predicts that the force decreases proportionally to the flap width and has a finite value ($\gamma t\cos\theta$) when the width tends to zero. This implies that near the tip, adhesion forces are negligible and the fracture force is the only remaining obstacle to detachment.

3. EXPERIMENTAL

To experimentally check equations (7) and (8), we have tested an adhesive film used as a window polarizer, of several thicknesses. These materials are brittle: they are easy to tear and on fracture leave behind two planar crack lips. Although the fracture force is different for each film, the adhesion energy is the parameter that can be more easily varied systematically. This can be done in two ways: by pulling the flap at different speeds or by using different substrates.

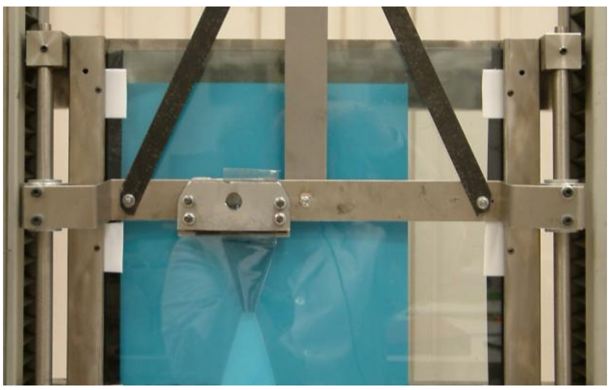

Figure 4. Experimental set-up.

The commercial adhesive film we used in the experiment was 3M Prestige 70 film, thickness 23-38 µm. The film was adhered to glass, steel and stainless steel plates, and parallel flaps 2-6 cm wide and of variable length was then cut and detached starting from the edge of the film. To include anisotropic effects, we cut and pulled flaps in the film in two perpendicular directions. The strip was then pulled with the help of a testing machine that leads to uniform pulling in all runs of the experiment, at speeds ranging from 80 down to 2 mm/min. A typical load-displacement record is shown is Figure 5.

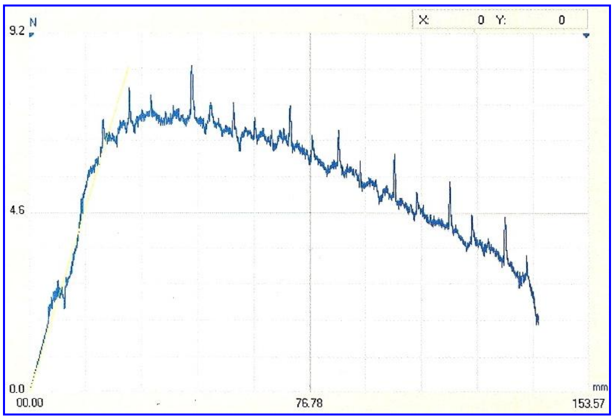

Figure 5. Load vs displacement recorded in a tearing-debonding test.

4. RESULTS AND DISSCUSION

The relation between adhesion energy and speed, $\tau = \tau(v)$, is not fully understood even though this dependence has been extensively studied in recent years [6-8]. As shown in Figure 6, by increasing the speed the adhesion energy increases and the tears become shorter.

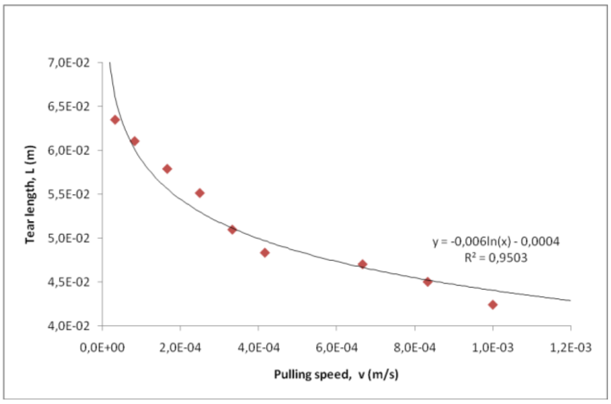

Figure 6. Tear length vs pulling speed for tearing-debonding tests.

In Figure 7, the pulling force, F, is plotted as a function of the flap width, W, for a number of experiments at a variety of pulling speeds, v. A linear decay of the force with the width of the tear is observed, as predicted by equation (7). The straight lines that fit each set of data have approximately the same intercept with the *y* axis. This is in accordance with equation (7), and thus the intercept gives an estimation of the fracture force. The value of the fracture force obtained in this way is consistent with the direct measurement of the force needed to start a tear by pulling a rectangular flap when the film is not adhered to a substrate, but held on its Boundaries (ASTM D1938-06) [9].

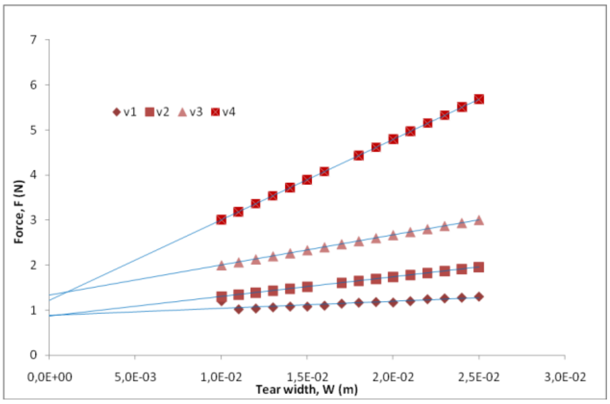

Figure 7. The pulling force versus flap width for runs made at different speeds.

The intercepts in Figure 7 are only an estimation of the fracture force since the straight lines intersect the y axis at $F = \gamma t\cos\theta$. Thus, the intercepts of those lines also depends on the tear angleθ. To obtain a better estimation of the fracture force, we extract the adhesion from the slopes in previous Figure 7 and plot the data using the modified equation:

$$\frac{F}{\cos\theta} = \frac{\tau W}{2\cos\theta} + \gamma t \qquad (9)$$

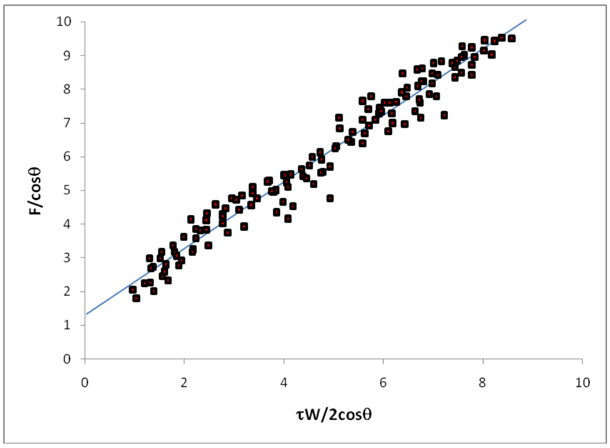

Figure 8. Estimation of the fracture force from the tearing-debonding test.

As we can expect, the slope for the best fit is ≈1. Let's now turn to equation (8). We need to obtain the exact expression for the elastic energy and, in general, it can be a difficult task to compute precisely how the elastic energy is distributed in the strip because the typical displacements observed are of the order of the system size. We take advantage of the film being strongly adhered to a substrate. This configuration helps to keep the lines across the flap width with zero curvature, allowing the surface to deform only along its longitudinal direction. The deflection can therefore be analyzed in terms of the classical elastica of Euler that accounts for arbitrary planar deformations of a sheet, and the elastic energy available for fracture can be easily obtained

$$U_E = \frac{4BW}{h} \tag{10}$$

$$\sin\theta = \frac{4B}{\gamma t h} \tag{11}$$

Figure 9 shows the experimental verification of equation (11). The variation of the angle and average distance h is produced by changing the substrate and varying the pulling speed. The solid line shows the theoretical prediction. The error bars show the uncertainty obtained from the estimated error of each parameter. We find the tear angle to be constant in our experiments (triangular tear shapes), this relation implies that *h is* a constant, throughout the tearing process.

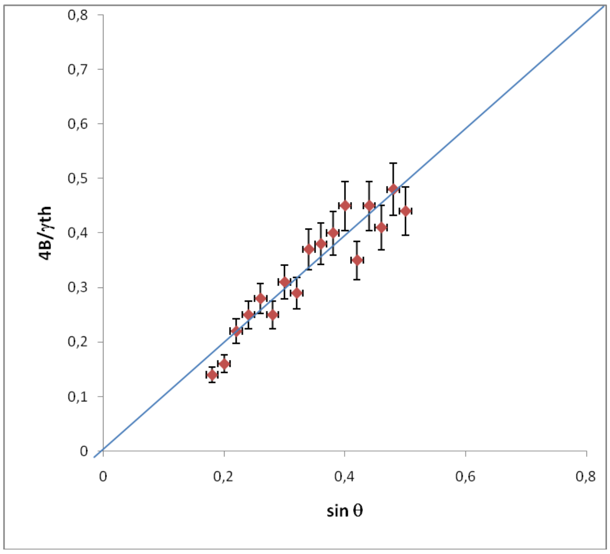

Figure 9. Experimental verification of equation (11).

So far, we have shown that relations (3) and (4) are satisfied by our experiments. It remains to be explained why these equations imply that the fracture trajectories are straight lines. This relation is consistent with the force measurements in Figure 10, but with a lower value of than the value η=1 expected for a perfectly elastic strip. The straight solid line with a slope η= 0.55 is the best fit for all of the experimental points and the dashed lines show the error bounds of our estimate. Equation (7) becomes now Equation (12)

$$4\eta^2 \frac{BW}{h^2} = \tau \frac{W}{2} + \gamma t \cos\theta \tag{12}$$

For large values of W, the last term in equation (12) is negligible, and the distance *h* must have the constant value

$$h = 2\eta \sqrt{\frac{2B}{\tau}} \tag{13}$$

Equation (12) shows that a larger pulling speed increases the adhesion energy, makes the fold joining the crack tips smaller (h decreases) and, the tears shorter.

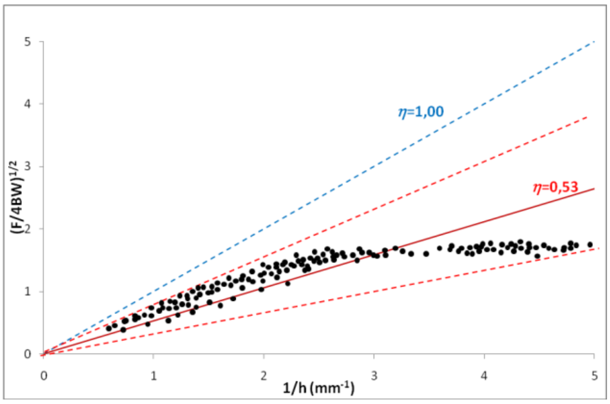

Figure 10. Value of the parameter η extracted from the experiments.

The trajectories are straight lines with a tear angle determined by three material constants: the elastic stiffness of the film, its fracture force and the adhesion energy with the substrate. When a flap is pulled to produce a tear, energy is localized in a narrow region connecting the flap with the film and becomes available for fracture. The specific geometry of the resulting fold gives a different elastic energy driving the fracture and lead to new tear shapes with no straight sides. Thus, under new conditions, different tear shapes can emerge that hide in their geometry the mechanism transforming elastic energy into surface energy of fracture and/or adhesion, and can potentially be used for mechanical characterization.

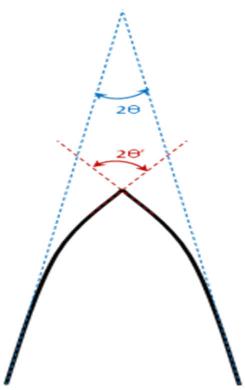

Figure 11. Deviation from straightness in the vertex of the flap.

5. CONCLUSIONS

1. A new method has been presented to investigate the mechanical properties of thin adhesive films. As thickness is reduced owing to new technologies,

traditional methods used to measure mechanical properties of a material in bulk form are not applicable, and leads to unexpected mechanical behaviour such as stress localization and wrinkling.

2. The coupling between elasticity, adhesion and fracture, imprinted in a tear shape, can be used to evaluate mechanical properties of thin films. The angle observed is a combination of three parameters: the elastic stiffness of the film, its fracture force and the adhesion energy with the substrate.

3. For a cylindrical deformation of the fold, η=1. A lower value of η observed in the experiments implies that the fold shows more rigidity than predicted by elasticity. Long interaction effects due to adhesive filaments also modify the shape of the fold.

4. The effective value of η is related to the cohesive law of the adhesive. Tear shapes hide in their geometry the mechanism transforming elastic energy into surface energy of fracture and/or adhesion, and can be used for mechanical characterization

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